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No. 4. A RADIOACTIVE GAS FROM CRUDE PETROLEUM, BY E. F. BURTON.

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BY

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In the course of their investigations on the radioactivity of the atmosphere Elster and Geitel[1] have shown that the soil and rock-masses constituting the surface layers of the earth are the source of an emanation, or gas, which gradually escapes into the air, and there exhibits properties analogous to the radioactive emanations from thorium and radium. In a conjoint paper by Professor McLennan and myself[2] on the conductivity of air confined in receivers of different metals some observations are cited which indicate that metals generally are, to a slight degree, the source of a similar emanation. This result has since been confirmed by Strutt,[3] who found that air drawn through a glass tube heated just below redness and containing scrap copper acquired a conductivity three or four times its normal value. Strutt[3] has also shown that a highly radioactive emanation can be obtained by bubbling air through mercury heated to about 300°C. More recently Professor J. J. Thomson[4] established the existence of a radioactive gas in the Cambridge tap-water, as well as in the water from a number of wells in different parts of England. Similar results have been obtained by Himstedt[5] at Freiburg, and by Lord Blythswood and H. S. Allen[6] with the mineral waters of Bath. Later still Adams[7] made a careful study of the radioactive gas in Cambridge tap-water, and his results, as well as those of Strutt on the emanation from mercury, go to show that the activity in all these cases is due to the presence of a substance very similar to, if not identical with, the emanation from radium.

In the following paper an account is given of some experiments with a highly radioactive gas obtained from crude petroleum, which, both in the rate at which its activity decays and

[1] Phys. Zeit., 3 Jahr. 24, p. 574. Denkschr. d. Kommission fur luftelect. Forschungen (München, 1903).

[2] Phil. Mag., 5th series, June, 1903, p. 699.

[3] Phil. Mag., 6th series, July, 1903, p. 113.

[4] Proc. Camb. Phil. Soc. xii, 3, 1903, p. 172.

[5] Berichte der Naturf. Ges. von Freiburg i. B., 1903, xiii, p. 101.

[6] Nature, Jan. 14, 1904, p. 247.

[7] Phil. Mag., 6th series, November, 1903, p. 563.

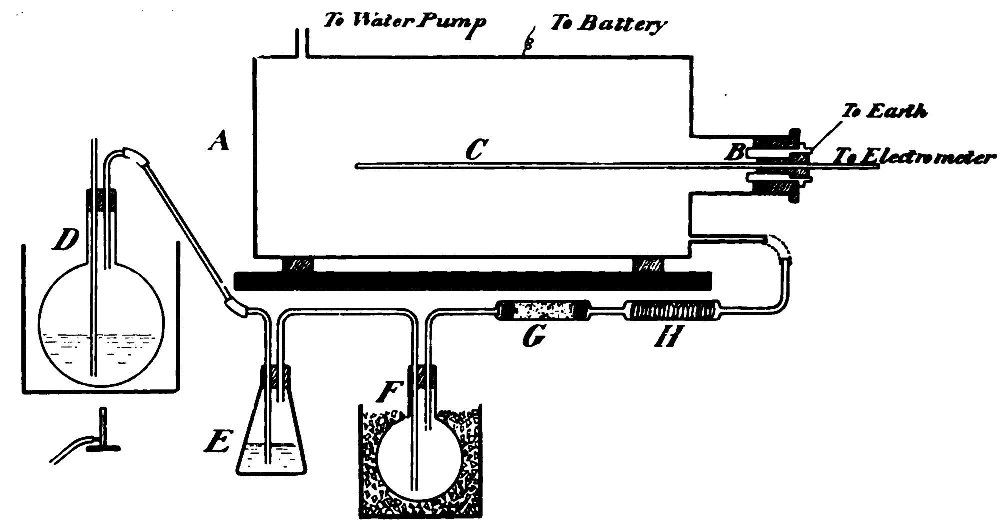
passing through the petroleum, becomes mixed with some radioactive gas or emanation. The initial portion of the curve leading up to the maximum corresponds exactly to that of the curve given by Rutherford[1] for the emanation from radium, and also to that of the curve given by Strutt for the radioactive gas obtained by bubbling air through mercury, and may be explained in the same way. The value of the conductivity immediately after the cylinder has been sealed measures the ionization due to the emanation itself. But, according to the disintegration theory proposed by Rutherford, the emanation is continuously producing by its decay the matter which causes excited radioactivity, and the ionizing power added by this latter material more than neutralizes, for a time, the decrease due to the decay of the emanation. Thus the conductivity of air freshly charged with this emanation gradually increases to a maximum state, which is reached when the loss in the ionizing power due to the decay of the emanation is just equalled by the gain contributed by the excited radioactivity produced in this process of decay.

From this time the rate of change indicated gives the rate of decay of the emanation. The law which the rate of decay of the emanation from radium follows may be expressed by the equation:

$$I_t = I_0 e^{-\lambda t}$$

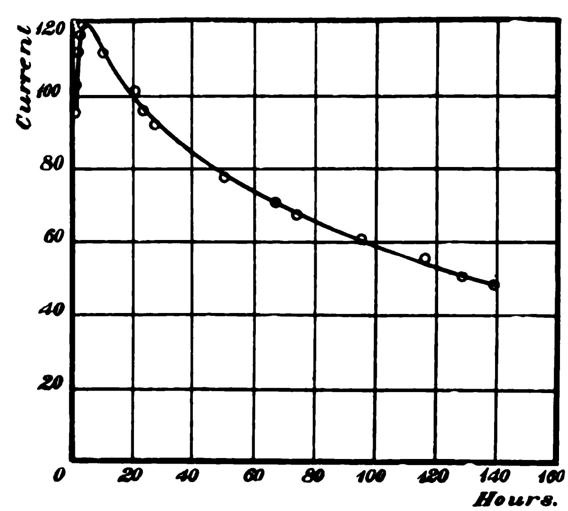
where I_0 is the value of the conductivity at any given time, I_t the value after an interval of t seconds, e the base of natural logarithms and λ a constant. By using this equation the values of $1/\lambda$ have been determined for a number of pairs of the readings given above and the results are tabulated in Column I of Table II. These values of $1/\lambda$, which give a mean of 557,000, show a marked increase with the time, and consequently indicate that the rate of decay is slower than that required by the law given above. This departure from the law of decay is probably due to a slight trace of a more persistent radioactive substance in the gas than the emanation and will be referred to later.

[1] Phil. Mag., 5th series, April, 1903, p. 445.



To Water Pump
To Battery
To Earth
To Electrometer
A
B
C
D
E
F
G
H

Fig. 1.



Current
120
100
80
60
40
20
0
20
40
60
80
100
120
140
160
Hours.

Fig 2.

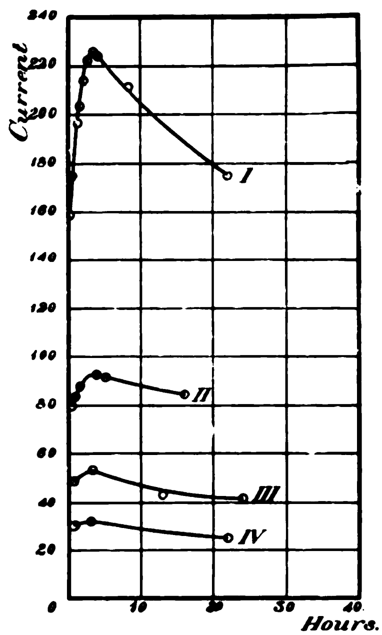

Current
240
220
200
180
160
140
120
100
80
60
40
20
0
10
20
30
40
Hours.
I
II
III
IV

Fig. 3.

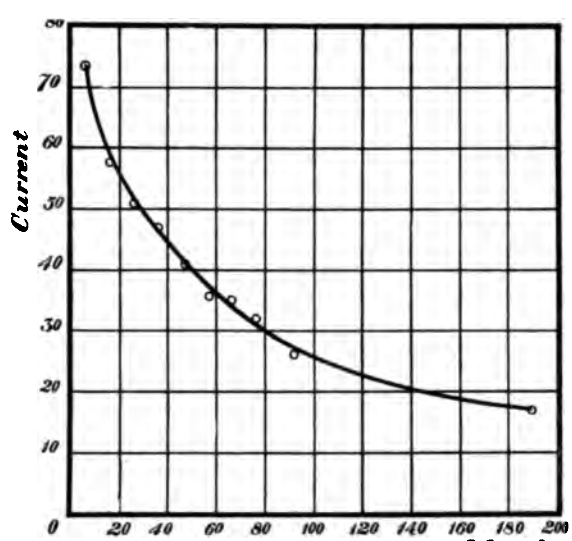

Current
70
60
50
40
30
20
10
0
20
40
60
80
100
120
140
160
180
200
Minutes.

Fig. 4.

TABLE II.

Column I. Burton.			Column II. Strutt.			Column III. Adams.		
Time in Hrs.	Current: Arb, Scale.	$\frac{1}{\lambda}$	Time in Hrs.	Current: Arb. Scale.	$\frac{1}{\lambda}$	Time in Hrs.	Current: Arb. Scale.	$\frac{1}{\lambda}$
0	119.7		0	140		0	188	
		360,000			379,000			366,000
17	101		18	118		16.7	160	
		414,000			389,000			401,000
47	77.8		42	94.5		40.4	129	
		669,000			472,000			494,000
64	71		66	78.7		64.8	108	
		617,000			504,000			381,000
92	60.3		90	66.3		88.9	86	
		726,000			371,000			372,000
135.6	48.6		140.5	40.6		139.6	53	
								573,000
						160.8	46	
$\frac{1}{\lambda}$ = 557,000			$\frac{1}{\lambda}$ = 423,000			$\frac{1}{\lambda}$ = 425,000		
Half value in 3.125 days.			Half value in 3.18 days.			Half value in 3 days.		

In Column II of Table II is given a set of Strutt's readings for the ionization due to the radioactive gas in mercury, and in Column III the values obtained by Adams with the active emanation in Cambridge tap-water. The calculated values of $^1/\lambda$ are inserted in both cases, but do not show the increase exhibited by the numbers in Column I. The averages of the three series of values of $^1/\lambda$ given in Table II, together with the mean values of the same constant obtained by Mme. Curie[1] and by Rutherford[2] for the decay of the emanation from radium, as well as the mean value calculated from Himstedt's results for the radioactive gas in water are collected in Table III. The values-show a very close agreement, and lead to the conclusion that the active gases from petroleum, spring water, and mercury are very probably identical with the emanation from radium.

1. Thèses prés. à la Faculté des Sci. de Paris, 1903.
2. Phil. Mag., 5th series, April, 1903, p. 445.

TABLE III.

Experimenter.	Source of Emanation.	Value of $\frac{1}{\lambda}$
Mme. Curie	Radium	497,000
Rutherford	Radium	463,000
Strutt	Mercury	423,000
Adams	Tap-water	425,000
Himstedt	Water	491,000
Burton	Petroleum	557,000

In his experiments with the water from the Cambridge mains Professor J. J. Thomson found that when the water had once been well boiled the gas expelled on any subsequent re-boiling was not appreciably radioactive. In the present investigation air was drawn through a selected sample of oil into the cylinder on three consecutive days and again on the sixth day, the first measurement being made about 24 hours after the petroleum had been pumped from the well. Each time the oil was used the bath was brought up to the boiling point and the air bubbled through it for 15 minutes, when observations on the conductivity of the air in the cylinder were commenced and continued at intervals over a period of about 20 hours.

TABLE IV.

Curve I.			Curve II.			Curve III.			Curve IV.		
Time. H.	M.	Current: Arb. Sc.	Time. H.	M.	Current: Arb. Sc.	Time. H.	M.	Current: Arb. Sc.	Time. H.	M.	Current: Arb. Sc.
	10	158.7		30	80		40	49		35	29.8
	30	174.2	1		83.4	3	10	53		55	30.3
1	5	196.7	1	30	87.2	13		43	3		31.6
1	30	203.7	3	40	92.6	24		41.5	22		25.2
1	50	214.2	5		92						
2	30	222.5	16		84.3						
3	30	226									
4		224.2									
8	15	211.1									
22		176									

The results, which are embodied in Table IV and illustrated by the curves in Fig. III, show that the activity acquired by

fresh air when drawn through the oil gradually decreased from day to day. The curves corresponding to the different tests exhibit the same characteristics as that in Fig. II. In each case the conductivity rose to a maximum in about three hours and then gradually decreased. The maximum currents in the four trials were respectively 13.9, 5.6, 3.2, and 1.9 times the conductivity of the ordinary air, thus showing that the oil at the end of a week still possessed in a marked degree the power to impart radioactivity to air drawn through it. Experiments made with a sample of oil which had been used in some preliminary tests and had been placed aside in a tightly corked glass vessel for over a month gave values almost identical with those represented by Curve IV, Fig. III, the maximum conductivity impressed in this case being 1.6 times that of the normal air. From these results it would appear that there is present in crude petroleum an active substance more persistent than the emanation from radium, perhaps a minute quantity of radium itself. If this be so, the air drawn through the oil might possibly carry with it into the cylinder a slight trace of this substance. Such a condition would explain the departure from the law of decay $I_t = I_0 e^{-\lambda t}$ exhibited by the increasing values of $1/\lambda$ in Column I of Table II.

Induced Radioactivity.—Each time the gas containing the emanation was blown from the cylinder the conductivity of the ordinary air admitted from the room was found to be still very high. Repeated tests showed that the initial conductivity of this fresh air was about 35 per cent. of that of the displaced gas, but in every case it quickly fell, until after about two hours the conductivity reached the normal value of 16.5. In expelling the emanation a blast of air was sent through the cylinder continuously for five minutes by means of a small foot-pump, after which the receiver was again sealed.

Measurements were then made on the conductivity at short intervals, and in Table V are given the results of one of these tests, the time being taken from the closing of the cylinder. In this particular case, the cylinder while filled with the air con-

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